

## \* NOTICES \*

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**CLAIMS**

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## [Claim(s)]

[Claim 1] $R_1R_2R_3R_4N^+$  and  $X_1^-$  ( $R_1$  in a formula,  $R_2$ ,  $R_3$ , and  $R_4$  an alkyl group of the carbon numbers 1-5 which are the same or are different) [ show and ]  $X_1^-$  -- Cl or Br -- being shown -- with the 4th class alkylammonium halide expressed.  $LiBF_4$  or  $LiPF_6$  is made to react in alcohol,  $R_1R_2R_3R_4N^+$  and  $X_2^-$  ( $R_1$  in a formula,  $R_2$ ,  $R_3$ , and  $R_4$  -- the above -- the same.)  $X_2^-$  --  $BF_4^-$  or  $PF_6^-$  -- being shown -- a method of manufacturing quarternary ammonium salt expressed.

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[Translation done.]

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## DETAILED DESCRIPTION

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### [Detailed Description of the Invention]

[0001]

[The technical field to which an invention belongs] This invention relates to the manufacturing method of the 4th class alkyl ammonium salt useful as an electrolyte of an electrolytic condenser or the organic electrolysis liquid of a cell.

[0002]

[Description of the Prior Art] Conventionally, the 4th class alkyl ammonium salt is manufactured by making the solution of the 4th class alkylammonium chloride, a star's picture, hydroxide and fluoroboric acid, or hexa fluorophosphoric acid react. Anhydrous hydrofluoric acid is used for a solvent and it is manufactured as indicated by JP,5-286981,A.

[0003]

[Problem(s) to be Solved by the Invention] Since the quaternary ammonium salt obtained in a conventional method has the large solubility to water, a yield is bad. For this reason, it condenses and the yield is raised. In such a case, many chlorine or bromine of a raw material cannot be contained, purity of a product cannot fall, and it cannot be used as an electrolytic condenser or an electrolyte. Therefore, recrystallization is repeated for high-grade-izing of a product. Since, as for the method of using anhydrous hydrofluoric acid as a solvent, there is a problem in handling and it cannot remove hydrogen fluoride thoroughly, it must be recrystallized with methanol. The issue which this invention tends to solve is canceling the difficulty of the above-mentioned conventional method, and is developing the method the object of a high grade being manufactured easily in more detail without using any recrystallization and anhydrous hydrofluoric acid.

[0004]

[Means for Solving the Problem] At this invention, they are  $R_1R_2R_3R_4N^+$  and  $X_1^-$  ( $R_1$  in a formula,  $R_2$ ,  $R_3$ , and  $R_4$  an alkyl group of the carbon numbers 1-5 which are the same or are different) in alcohol. [ show and ]  $X_1^-$  -- Cl or Br -- being shown -- with the 4th class alkylammonium halide expressed. making  $LiBF_4^-$  or  $LiPF_6^-$  react --  $R_1R_2R_3R_4N^+$  and  $X_2^-$  ( $R_1$  in a formula,  $R_2$ ,  $R_3$ , and  $R_4$  -- the above -- the same.)  $X_2^-$  --  $BF_4^-$  or  $PF_6^-$  -- being shown -- it is making to manufacture quaternary ammonium salt expressed into the gist.

[0005] An alkyl group of quaternary ammonium salt of this invention has a carbon number as small as 1-5, and points out methyl, ethyl, n-propyl, iso-propyl, n-butyl, iso-PUCHIRU, t-butyl, n-pentyl, iso-pentyl, and t-

penty. An alkyl group of quaternary ammonium salt consists of one kind of thing in the above-mentioned alkyl group, a thing combined two kinds, or two or more kinds of combination.

[0006]Alcohol of a solvent to be used is lower aliphatic alcohol, such as methanol, ethanol, n-propyl alcohol, and iso-propyl alcohol, and is methanol and ethanol preferably. It may heat, although a reaction is usually performed at ordinary temperature.

[0007]

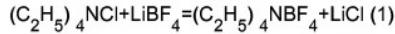
[Embodiment of the Invention]this invention method is a method of obtaining the product of a high grade using the solubility difference over alcohol of each salt. The solubility (25 \*\*) to alcohol was shown in Table 1.

[0008]

[Table 1]

| 塩  | 溶解度 (g/100 sat) |       |
|--|-----------------|-------|
|  | メタノール           | エタノール |
| LiCl   | 30.5            | 20.2  |
| LiBr   | 58.2            | 41.4  |
| (C <sub>2</sub> H <sub>5</sub> ) <sub>4</sub> NBF <sub>4</sub>   | 7.5             | 4.3   |
| (C <sub>2</sub> H <sub>5</sub> ) <sub>4</sub> NPF <sub>6</sub>   | 4.3             | 2.3   |
| (n-C <sub>3</sub> H <sub>7</sub> ) <sub>4</sub> NPF <sub>6</sub> | 3.2             | 6.6   |

[0009]The reaction formula at the time of making a tetraethylammonium chloride and lithium borofluoride react in methanol was shown in (1).



Target ammonium salt and lithium chloride generate. However, since the solubility of ammonium salt is small, a product deposits out of a solution, but since the solubility to methanol of a lithium chloride is large, it does not deposit from a reaction solution. Therefore, the dissolution is separated and the product of a high grade is obtained only by washing a crystal and drying. Thus, the obtained product remains as it is and it is usable as an electrolyte of electrolysis KONDESA or a cell in it. In order to raise purity, it may recrystallize in alcohol.

[0010]

[Example]While the typical example of this invention is shown below, it explains still more concretely. These are mere illustration for explanation and it cannot be overemphasized that this invention is not what is restricted to these in any way.

[0011]

[Example 1] The tetraethylammonium chloride 165.7g (1 mol) is dissolved in 500 ml of methanol. On the other hand, the lithium borofluoride 93.8g (1 mol) is dissolved in 500 ml of methanol. Both solutions are mixed and it agitates for 30 minutes. The crystal of the Howe tetraethylammonium fluoridation deposits. Although the lithium chloride which carried out the byproduct is 42.4g, the solubility to this thing methanol is 30.5g / 100g sat, and dissolves not less than 300g to 1000 ml of methanol. Therefore, a lithium chloride does not deposit from a solution. The solution was \*\*\*\*(ed), 100 ml of methanol washed the crystal 3 times,

and it dried at 100 \*\* among the nitrogen air current for 48 hours. The purity of 151.5 g (70% of yield) and a product of the yield of the product was 99.9%, and moisture was 20 ppm or less. Impurities were Fe 1ppm max and Na 2ppm.

[0012]

[Example 2] The tetraethylammonium chloride 165.7g (1 mol) is dissolved in 500 ml of ethanol. It dissolves cooling the lithium 151.9g (1 mol) hexafluorophosphorate to 500 ml of ethanol. After \*\*\*\*(ing) a solution, it adds in the solution of a tetraethylammonium chloride, and it agitates for 30 minutes at ordinary temperature. The crystal of tetraethylammonium hexafluorophosphorate deposits. Since the lithium chloride which carries out a byproduction is dissolving thoroughly into ethanol, it does not deposit. After \*\*\*\*(ing) a solution and washing a crystal 3 times by 100 ml of ethanol, it was made to dry at 100 \*\* among a nitrogen air current for 48 hours. The purity of 239.4 g (87% of yield) and a product of the yield of the product was 99%, and moisture was 100 ppm or less. Impurities were less than Fe 1ppm and less than Na 1ppm.

[0013]

[Example 3] The tetra-n-butylammonium bromide 322g (1 mol) is dissolved in 500 ml of methanol. LiPF<sub>6</sub> 151.9g (1 mol) is dissolved in 500 ml of methanol, and it adds in a previous solution. It agitates at ordinary temperature for 1 hour. The lithium bromide which carries out a byproduction is 87.9g, and dissolves in 1000 ml of methanol enough. The crystal was \*\*\*\*(ed) and it was made to dry at 100 \*\* among an after-washing nitrogen air current with methanol for 48 hours. 329g (85.1% of yield) of tetrahexafluorophosphorate n-butyl ammonium was obtained. The purity of the product was 99% and moisture was 100 ppm or less. Impurities were less than Fe 1ppm and Na1ppm.

[0014]

[Effect of the Invention] Since the solubility difference to the inside of alcohol of the reactional solvent of a resultant is used, moreover, the purpose products can be easily obtained by a high grade.

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[Translation done.]